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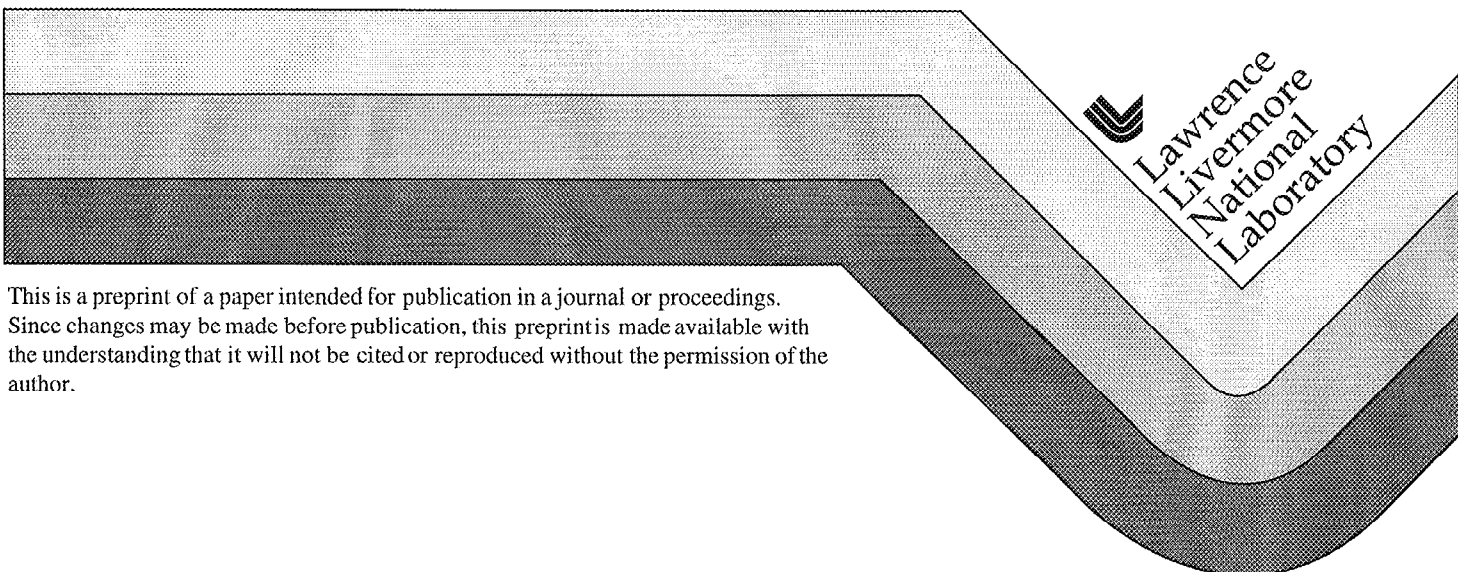
PREPRINT

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# Application of CZT detectors in nuclear materials safeguards

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## ABSTRACT

High-resolution, gamma- and X-ray spectrometry are used routinely in nuclear materials safeguards verification measurements. These measurements are mostly performed with high-purity germanium (HPGe) detectors, which require cooling at liquid-nitrogen temperatures, thus limiting their utility in field and unattended safeguards measurement applications. Sodium iodide (NaI) scintillation detectors do not require cooling, but their energy resolution (10% at 122 keV) is insufficient for many verification measurements. Semiconductor detectors that operate at room temperatures, such as cadmium-zinc-telluride (CZT) detectors, with energy resolution performance reaching 2.0% at 122 keV may be used for certain safeguards verification applications. We have developed hardware to utilize CZT detectors in X- and gamma-ray measurement systems and software to apply such a system in measuring  $^{235}\text{U}$  enrichment for safeguards verification purposes. The paper reports on the CZT detector-based measurement system and measurement results obtained with it. The paper also discusses work on additional improvements to broaden the applications of the system.

**Keywords:** CZT detector, gamma-ray spectrometry, uranium enrichment, verification, nuclear material safeguards

## 1. Introduction

Nuclear materials safeguards are measures to guard against loss or diversion of nuclear materials from permitted uses, and to give timely indication of possible diversion or credible assurance that no diversion has occurred. Most materials of concern in nuclear safeguards emit  $\gamma$ - and X-rays and these can be used for nondestructive assay (NDA) of the materials to verify nuclear material inventories.

The  $\gamma$  and X rays have well defined energies which are characteristic of the isotopes emitting them. Their intensities are used to determine the isotopic composition of the materials and can also be used to provide quantitative information on the amount of material present. Enriched uranium fuel, for example, has a strong 186 keV  $\gamma$  ray associated with the  $\alpha$  decay of  $^{235}\text{U}$  and the  $^{235}\text{U}$  enrichment can be verified by measuring the intensities of this  $\gamma$  ray. Plutonium samples generally contain the isotopes of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{242}\text{Pu}$  along with decay products, which gives rise to a complex spectrum of characteristic  $\gamma$  and X rays. The intensities of these emissions are used to determine the isotopic composition of the plutonium and whether it is weapons-grade. The date of discharge of irradiated fuel from a reactor can be verified by measuring the relative intensities of  $\gamma$  rays associated with fission and activation products. The 662 keV  $\gamma$  ray from  $^{137}\text{Cs}$  is particularly important for this type of measurement.

NaI detectors can be made with large volumes and generally have higher  $\gamma$  ray detection efficiencies than HPGe detectors. Their uses in safeguards applications include the verification of fresh  $^{235}\text{U}$  fuel enrichment as well as the presence of spent fuel through the detection of fission product  $\gamma$  radiation. Their ability to distinguish between  $\gamma$  rays of different energies is relatively poor. The  $^{235}\text{U}$  enrichment measurement technique with NaI detectors requires calibration and also that the measured items meet the calibration conditions. This technique cannot be used to measure arbitrary samples of uranium.

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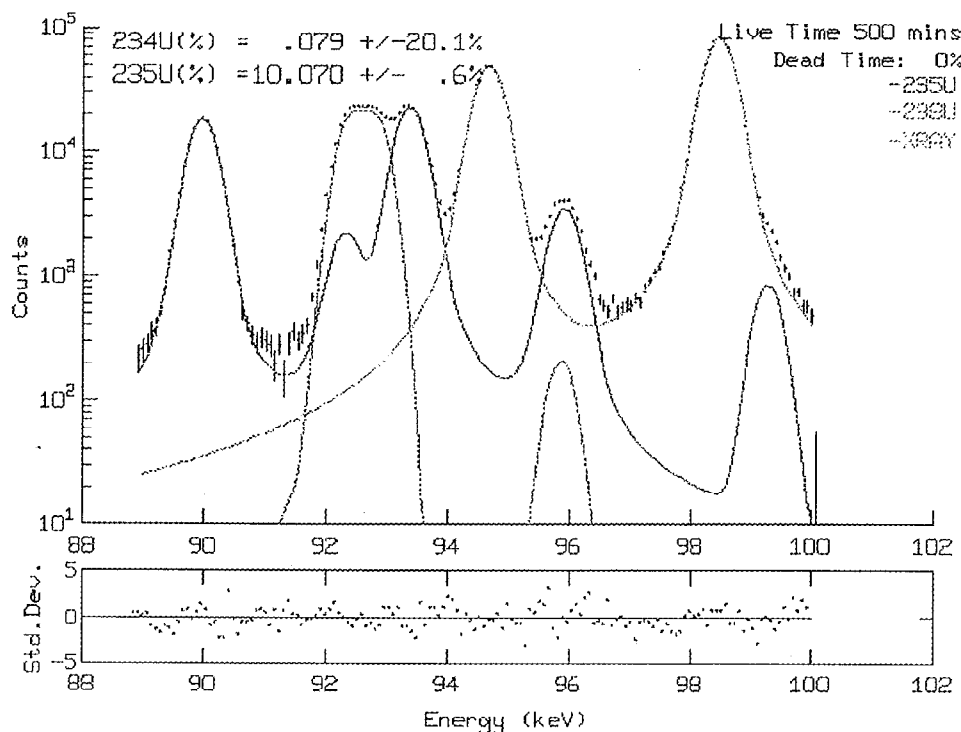


Figure 1. This plot of the 88- to 100-keV region of a uranium spectrum with a  $^{235}\text{U}$  enrichment of 10% obtained with a high-resolution germanium detector shows the component responses for  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and U X-rays.

Germanium detectors have much better energy resolution than NaI detectors and are much better suited to resolving complex  $\gamma$ -ray spectra and providing information about the isotopic composition of nuclear materials. An example is a  $\gamma$ -ray measurement method to determine  $^{235}\text{U}$  enrichments in items where no suitable calibration standards exist or where non-reproducible conditions make calibration impossible. A  $\gamma$ -ray analysis technique has been developed to determine  $^{235}\text{U}$  enrichments by analyzing the  $\gamma$ - and X-ray peaks in the 88- to 100-keV region of a uranium spectrum.<sup>1</sup> An advantage in analyzing this region is the  $^{235}\text{U}$  and  $^{238}\text{U}$  photon emissions are very close in energy and therefore detected with comparable efficiencies. This analysis approach eliminates the need for calibration and allows  $^{235}\text{U}$  enrichment measurements of arbitrary samples. The complex structure of this region with closely overlapping peaks is the greatest obstacle to accurate analyses. To achieve high accuracy and precision, this analysis requires good energy resolution as can be seen in Figure 1 in which the spectral data were measured with a small, planar germanium detector with energy resolution of 0.5% at 122 keV.

HPGe detectors have limited utility in field and unattended safeguards measurement applications, because they require cooling to liquid nitrogen temperatures. NaI detectors do not require cooling, but their energy resolution is insufficient for this uranium enrichment measurement technique. We have developed hardware and software that allows use of carefully selected CZT detectors to perform these measurements with a battery-operated, completely portable measurement system.

## 2. System Hardware

A completely portable CZT detector system is shown in Figure 2. It consists of a laptop computer that contains both the system operation and uranium enrichment analysis software, a portable multi-channel analyzer (MCA), and a CZT detector probe with electronics designed for CZT detectors.

The CZT detector electronics were designed specifically to optimize the CZT detector performance. Thorough consideration was given to all the CZT detector characteristics that contribute to less than ideal energy resolution including baseline noise (primarily shot noise), poor hole-mobility and charge trapping. The preamplifier and filter amplifier were

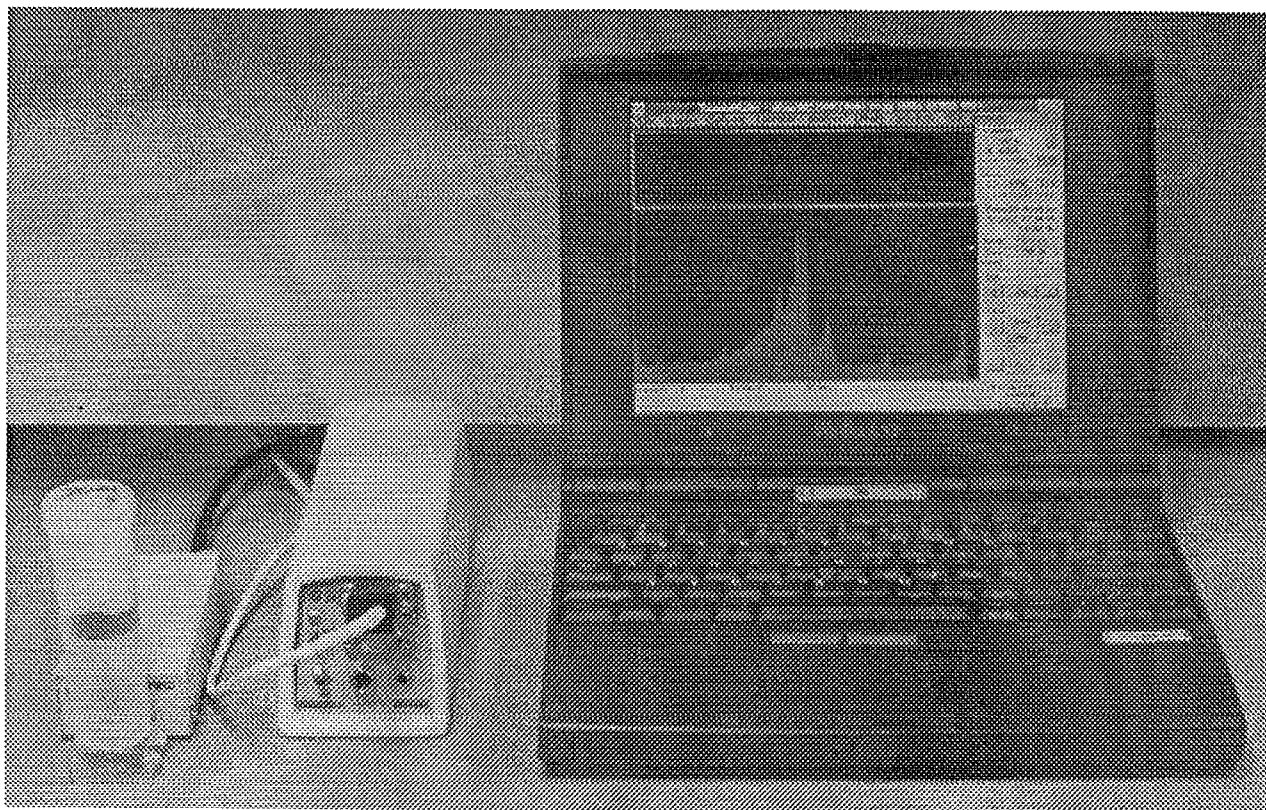


Figure 2. A portable CZT detector system with a laptop computer to control data acquisition and perform the uranium enrichment analysis, a portable multi-channel analyzer, and a CZT detector with specifically designed supporting electronics. The CZT detector with its electronics is on the left behind the sample being measured. A uranium spectrum can be seen on the computer display screen.

designed to minimize these effects and include monolithic devices, which allowed the current implementation of surface-mount technology for further size reduction. This design achieves excellent reduction of the spectral noise. The energy resolution of the 122 keV peak approaches 2.5 keV FWHM, when using the highest quality, spectrometry-grade 5x5x5 mm detectors. This monolithic design was able to achieve excellent resolution (10% better than commercial instrumentation) and low-power consumption (600 mW), which can be powered by a battery-operated MCA. A detailed description of the development of the CZT detector electronics is given elsewhere.<sup>2</sup>

### 3. System Software

The general method for determining the relative isotopic abundances from  $\gamma$ -ray spectra is to measure and interpret the  $\gamma$ -ray and X-ray peak intensities associated with the different isotopes. Relative isotopic abundances can be calculated with known branching intensities and half-lives for the isotopes and applying corrections for the differences in the detection efficiencies of the measured peaks. The relative efficiency factors can usually be determined from the measured spectrum, if the peaks are close in energy.

When the peaks severely overlap each other, the determination of the peak intensities becomes more complicated; then, peak-fitting techniques are required. Sometimes the overlap is so severe and the energy resolution is such that special steps must be taken in the fitting process so that proper convergence of the fit is obtained. In such circumstances, not all of the peak positions and heights can be allowed to be independently free in the fitting process. To overcome these difficulties, we have developed a "response function" method that fixes the peak energies and heights relative to each other for a given

isotopic component. We also fix other peak parameters like those describing the low-energy exponential tail used in our model of  $\gamma$ - and X-ray peaks. These parameters can be determined from other isolated peaks in the spectrum. This model and the fitting methods described here are incorporated into a computer program called CZTU.

We have applied these techniques to peaks in the 87-102 keV region of the uranium spectrum. In this region there are  $\gamma$ - and x-rays from three different sources: the decay of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and the U X-ray peaks due to self-induced fluorescence of the uranium sample. The net peak intensities in a given energy region are found by finding and subtracting a background function from the data. The background function in a spectrum acquired with a CZT detector is more difficult to accurately define than for germanium detectors. Because the CZT detectors being used are small and thus have poor detector efficiency, the measured spectral data are statistically poor. In addition, the  $\gamma$ - and X-ray peaks from CZT detectors all have relatively long low-energy tails that make finding the "correct" background more difficult. Both of these complications were addressed in the CZTU code by choosing as "good" a background as possible and then iterating other background choices around this initial value to see which background gave the best fit to the data.

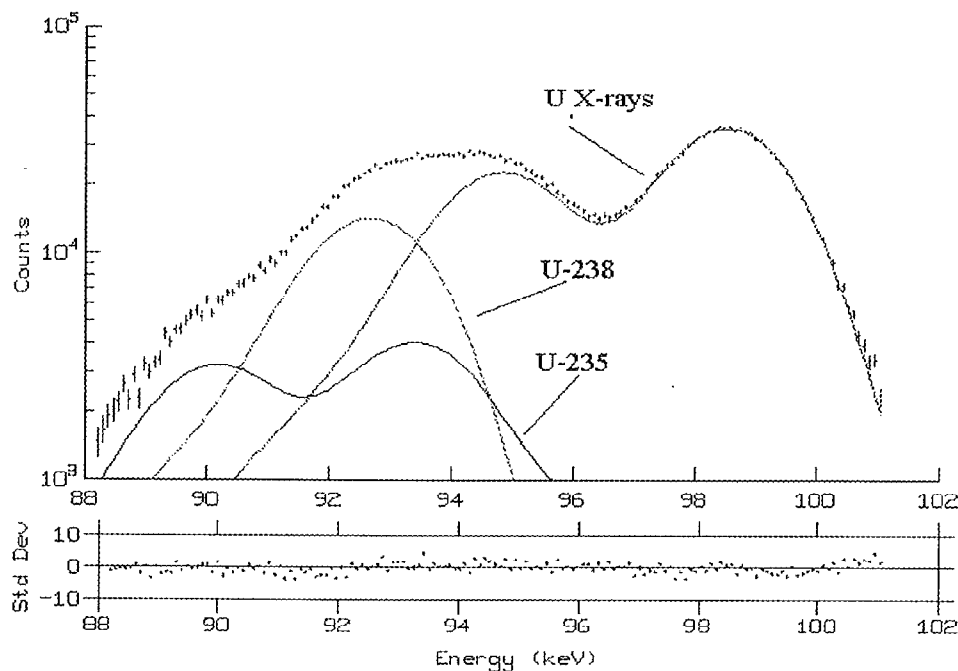


Figure 3. This plot shows an analysis similar to that shown in Figure 1 except the data were collected with a 5x5x5 mm CZT detector with an energy resolution of 2.5 keV at 122 keV. The enrichment of the uranium sample in this measurement was 4.46%.

We fit the resulting net data with "response profiles" for the  $^{235}\text{U}$ ,  $^{238}\text{U}$  and the U X-ray components. Note that  $\gamma$ - and X-ray peaks are different and require different peak-shape algorithms, which are incorporated into the CZTU program. Gamma-ray peak shapes with CZT detectors are described very well by a Gaussian peak and a low-energy exponential tail.<sup>3</sup> X-ray peak shapes are described very well by a convolution of the detector response (a Gaussian and low-energy exponential tail) with the intrinsic x-ray energy distribution (a Lorentzian)—a Voigt profile. A fit to the net data is shown in Figure 3.

#### 4. Measurement Results

To evaluate the applicability and accuracy of this method on uranium spectrum acquired with CZT detectors, we participated in an uranium enrichment exercise at the Institute of Reference Materials and Measurements in Geel, Belgium. A comparison of the results between results obtained with a CZT detector and those obtained with a high-resolution

germanium detector are shown in Table 1. The CZT detector used was a 5x5x5 mm device mounted with a prototype of our CZT detector electronics.

Sample X and Sample Y were fresh fuel; therefore, the  $^{238}\text{U}/^{234}\text{Th}$  non-equilibrium was corrected for during the analysis. With a few exceptions the results obtained with the CZT detector tracked the germanium detector results and were within the design specifications of  $\pm 10\%$ . We note that the counting times for the CZT detector were roughly twice that of the germanium detector.

Table 1  
Uranium Enrichment Measurement Exercise Results

Sample Number	U-235/U Certified (weight %)	HPGe (weight %)	Counting Time (min)	CZT (weight %)	Counting Time (min)
1613	$3.1094 \pm 0.0044$	$3.10 \pm 0.02$	75.5	$3.10 \pm 0.34$	133.4
1614	$1.4972 \pm 0.0019$	---	---	$1.73 \pm 0.20$	166.6
1541	$1.9952 \pm 0.0037$	$1.99 \pm 0.01$	61.9	$2.01 \pm 0.23$	161.4
1542	$2.8774 \pm 0.0040$	$2.87 \pm 0.02$	92.2	$2.84 \pm 0.32$	145.9
Sample X	$3.4317 \pm 0.0032$	$3.3 \pm 0.03$	75.4	$3.03 \pm 0.34$	158.8
Sample Y	$2.6846 \pm 0.0031$	$2.55 \pm 0.02$	67.8	$2.42 \pm 0.28$	179.7
125V	$93.1556 \pm 0.0227$	$89.39 \pm 4.87$	90.9	---	---

## 5. Summary

A truly portable, ambient-temperature, radiation measurement instrument that provides for uranium enrichment determination in the field or in unattended applications has been developed. While the accuracy of the enrichment measurements meet requirements, the long counting times required limit its use. There is a need for larger volume detectors with greater detection efficiency that have energy resolution performance similar to the 5x5x5 mm CZT detectors used here.

Digirad, a CZT manufacturer located in San Diego, California that specializes in applying CZT detectors to medical imaging, has developed a new CZT detector technology that provides dramatic improvement to the peak-to-valley spectral characteristics of CZT detectors. We have evaluated several new prototype detectors from Digirad (5x5x5, 4x4x4, and 3x3x3 mm) that provide a peak-to-valley of up to 60 to 1 at  $^{137}\text{Cs}$  (662 keV). Current detectors only provide a peak-to-valley of up to 7 to 1, with low-energy tailing being the limiting factor. This dramatic increase is due to the virtual elimination of low-energy tailing as a result of unique and proprietary single-charge collection techniques. While the energy resolution performance of CZT detection systems has been shown to be satisfactory for many nuclear safeguards applications, low detector efficiency remains the primary disadvantage. To address this issue, we are currently working with Digirad to develop large area detectors by implementing these detectors into arrays to improve counting efficiency, thus reducing counting times.

## 6. Acknowledgements

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## 7. References

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